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THEORY OF HIGH INTENSITY MULTIMODE
GAS LASERS - INCLUSION OF COLLISIONAL EFFECTS

FINAL REPORT

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13. ABSTRACT

Several topics in laser theory and the interaction of radiation with matter have been addressed. First, a mathematical theory of a high intensity single mode gas laser has been developed. New computational techniques have been applied which serve to unite several previous approaches to single mode laser theory. A generalized dressed atom approach was formulated to provide a new way of viewing the physical processes occurring when high intensity fields interact with matter. Second, a theoretical study of the manner in which laser spectroscopy can be used to probe collision effects in atomic and molecular systems has been undertaken. Both two and three level systems were studied; steady state and coherent transient techniques were examined. It is now apparent that laser spectroscopy will be a useful tool for the determination of various excited state differential and total cross sections.

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Coherent Transients						
Photon Echo						
Resonance Fluorescence						
Three-level Systems						
Pulse Propagation						

FINAL REPORT

1. Statement of Problem

The problem as stated in the original research proposal was to
develop a theory of high intensity multimode gas lasers and to incorporate
collision effects into the theory wherever possible. In practice, the
laser theory and collision problems were studied separately. In the area
of laser theory, we sought to connect previous theories of a high intensity
single mode gas laser and to provide new computational methods for solving
that problem which could be generalized to multimode theory. We also
sought a better understanding of the physical processes occurring in high
intensity lasers. Our studies of collision effects in atomic and molecular
systemswere strongly influenced by the rapid advances that were occurring
in Doppler-free laser spectroscopy. Our goal in this area became to
formulate methods by which one could use laser spectroscopic techniques
to obtain both differential and total cross section data for the ground
and excited states of atoms or molecules. This information could then
be used in the master equations of laser theory; in this way, one could
incorporate collision effects into laser theory. While working in the
area of laser spectroscopy, we were led to study problems related to re-
sonance fluorescence and laser propagation effects.

2. Content of the Report

The technical results obtained under this Grant for the period
15 August, 1973 - 30 June, 1976, have already been described in previous
Progress Reports (in particular, the Progress Report for the period 1
October, 1975 - 30 June, 1976, was quite extensive). Moreover, almost
all of the research carried out under this Grant has been or is in the
process of being published in readily available technical journals.
Consequently the remainder of this report will consist of (1) A summary
of the research carried out for the period 1 July, 1976 - 31 May, 1977,
(2) A brief summary of the major results obtained over the period of

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the Grant, (3) a list of personnel involved in the project, and (4)
a list of publications associated with the research carried out under
this Grant.

3. Summary of Research Findings for the period 1 July, 1976 - 31 May, 1977

Our study of the effects of collisions on the line shapes asso-
ciated with two and three level atomic and molecular systems has been more
fully developed. In an earlier publication[1*], we had used a phenomeno-
logical propagator to account for the way in which collisions change the
velocity associated with the populations of the various levels of the
atomic system under consideration. We have been able to re-solve the problem
using a somewhat more conventional and realistic propagator based on the
Keilson-Storer collision kernel (see ref. 1). In doing so, we now have a
theory that is valid over the entire range from "weak" to "strong" velocity-
changing collisions.

The physical system under investigation consists of either two
or three level atoms subject to two external laser fields and collisions
with perturber atoms. One laser field (pump) is in near resonance with
one of the transitions in the atoms and can excite a specific velocity
ensemble of the atoms. The second laser field (probe) is in near re-
sonance with the same or a coupled transition and probes any changes in the
velocity that have occurred as a result of collisions. The line shapes in-
volved are also sensitive to the effects of "phase-interrupting" collisions
which, in turn, are related to the total elastic scattering cross sections
for the various levels. Thus the line shape can serve to monitor differ-
ential (velocity-changing) and total cross sections. Both steady-state
and transient type experiments were analyzed.

A simple, but not unreasonable collision model was adopted, in
which collisions where "phase-interrupting" in their effect on off-diagonal
density matrix elements and "velocity-changing" in their effect on diagonal
density matrix elements. Within this model, we showed how to systematically

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*Superscripts refer to references in Section 6 of this report.

extract cross section data from the probe absorption line shape by using different pump detunings. The calculation was carried out in perturbation theory in the laser fields, but no assumption on the ratio of the detunings and widths to the Doppler width was employed. Predictions of the theory include the existence of the well known narrow saturated absorption resonance and a background "shoulder" due to velocity-changing collisions. It was also pointed out that the narrow resonance can exhibit a width with a nonlinear pressure dependence at low pressures. In a collaborative effort with the group at Orsay, France, experimental verification of various aspects of the theory has started to appear.[o]

Current research involves explicit numerical evaluations of the line shapes appearing in reference 1 and an explanation for the various types of structure that appear in those line shapes. Moreover, we are exploring the consequences of different collision models. For example, if the collision interaction is the same for two or three of the levels of the atoms, then collisions will be "velocity-changing" in their effect on off-diagonal density matrix elements. This type of model will lead to the possibility of line narrowing with increased pressure.

4. Brief Summary of Research Results Obtained Under the Grant

    . A. High Intensity Laser Theory - New computational techniques for studying the problem of a high intensity single mode gas laser were developed.[n] Among these were a backward recursion method, an integral equation approach and direct numerical solution of the differential equation with periodic boundary conditions. Such methods should be useful in the multimode problem where conventional continued fraction techniques can not be easily applied. We were also able to connect several previous theories of the high intensity laser, providing an overall unity to these theories.

    Additional insight into the physical processes occurring when two or more high intensity fields interact with atoms was obtained in a Generalized Dressed Atom Approach (GDAA). In the GDAA, we used part of

each field to "dress" the atoms and considered the interaction of the remaining interaction ("probe") with these dressed atoms. In this model we were able to semi-quantitatively predict to the position of resonances which occur in graphs of the spatially averaged population inversion in a standing wave gas laser as a function of axial atomic velocity. Such structure had been noted in previous theories, but not explained on physical grounds. As a related calculation[1], we were able to predict the position for multiphoton Bloch-Siegert resonances, having demonstrated the mathematical equivalence of the traveling wave stationary atom Bloch-Siegert and the standing wave moving atom laser problems. It is hoped that the GDAA will be applied to more complex systems to provide physical insight into those systems.

B. <u>Collision Effects in Atomic and Molecular Systems</u> - Many new and significant results were obtained in this area. A basic theory of collision effects in atomic and molecular systems[d,e] was applied to a number of experimental situations. In certain cases the theory was used to explain experimental results while in other cases the theory was developed in a fashion that suggests new types of experiments which could be carried out.

The theory was used to explain the intensity of photon echoes as a function of pulse delay for vibrational transitions in CH_3F.[a,f] Since the transition levels in CH_3F experience almost equal collision interaction with perturber atoms, a model of velocity-changing collisions was assumed. Such a model lends to an echo amplitude that varies exponentially as $-t^3$ for small pulse separations t and as $-t$ for longer t, in agreement with experiment. The photon echo experiment is the most direct evidence to date for velocity-changing collisions affecting <u>off-diagonal</u> density matrix elements. From the data, we obtained the cross section for velocity-changing collisions and the mean change in velocity per collision. This is the first experiment where both of these parameters could be determined.

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In working on photon echo theory, we also established the validity conditions for using the Fokker-Planck equation as a weak collision limit of the Boltzmann equation.[b,e] Especially in problems involving both radiation and collisions or in problems involving initially narrow velocity distributions, one can not arbitrarily take the Fokker-Planck limit of the Boltzmann equation, as has been commonly done.

Of considerable interest has been the study of collision effects in two and three level atomic systems subject to radiation fields.[i,m] One field (pump) excites a specific velocity ensemble of atoms on a given transition and a second field (probe) probes the same or a coupled transition. By monitoring the probe absorption as a function of probe detuning for various pump detunings, one can obtain information on the collisional processes occurring in the various levels. The line shapes can be used in two fashions. First, systematic line shape studies can be used to extract data on both the differential and total cross section for the various levels of the transitions. Thus, excited state cross section data should be readily available for the first time. Second, the line shapes can be used to check the validity of various theories of collision effects that have been recently proposed.[e] We believe that, in the next few years, nonlinear optical techniques of the type described above will find an ever increasing application in the determination of collision cross sections.

C. <u>Resonance Fluorescence</u> - A first principle calculation of resonance fluorescence for moving atoms excited by a monochromatic laser was carried out. The frequency spectrum and fluorescence distribution were determined, supportion the notion that the laser absorption and subsequent spontaneous emission must be looked at as a single process. An experiment to measure the atomic recoil in fluorescence was proposed.

D. <u>Self-Trapping of Laser Beams</u> - A moment-theoretical approach to the theory of self trapping of lasers in plasmas was developed.[k]

The approach led to a simple formulation in which the effects of saturation, the stability of propagation, and the effects of an active medium could be explored. It was found that a minimum threshold power was needed for stable beam propagation.

5. Personnel

        Paul R. Berman - Principal Investigator

        Jehuda Ziegler - Graduate Student and Assistant Research Scientist (Ph.D. Thesis - Theory of a High Intensity Gas Laser, October, 1975).

        Juan F. Lam - Graduate Student and Assistant Research Scientist (Ph.D. Thesis - A Dynamical Theory of Stationary Self-Focusing of Laser Beams, June, 1976).

6. Publications

a. R.G. Brewer, J. Schmidt, and P.R. Berman, "Coherent Transient Study of Velocity-Changing Collisions", Physical Review Letters 31, 1103 (1973).

b. P.R. Berman, "Brownian Motion of Atomic Systems: Fokker-Planck Limit of Transport Equation", Physical Review A9, 2170-2177 (1974).

c. P.R. Berman and J. Ziegler, "Dressed Atom Picture of High Intensity Gas Laser", Bulletin of American Physical Society 20, 636 (1975).

d. P.R. Berman, "Effects of Phase-Interrupting and Velocity-Changing Collisions on Spectral Line Formation", Comments on Atomic and Molecular Physics, 5, 19-25 (1975).

e. P.R. Berman, "Theory of Collision Effects on Atomic and Molecular Line Shapes, Applied Physics 6, 283-296 (1975).

f. P.R. Berman, J. Levy and R.G. Brewer, "Coherent Optical Transient Study of Molecular Collisions: Theory and Observations", A11, 1668-1688 (1975).

g. P.R. Berman and J. Ziegler, "Dressed Atom Picture of High Intensity Gas Laser", in Laser Spectroscopy edited by S. Haroche, J.C. Pebay-Payroula, T.W. Hänsch, S.E. Harris (Springer-Verlag, Berlin, 1975) p. 464.

Publications - Con't.

h. P.R. Berman "Theory of Collisions in Doppler-Free Spectroscopy", Physical Review A13, 2191-2211 (1976).

i. P.R. Berman, "Theory of Collision Effects in Doppler-Free Spectroscopy", Optics Communications 18, 1 (1976).

j. J.F. Lam and P.R. Berman, "The Effect of Recoil in Resonance Fluorescence," Phys. Rev. A14, 1683-1688 (1976).

k. J.F. Lam, B.A. Lippmann and F. Tappert, "Self-Trapped Laser Beams in Plasmas", to appear in Physics of Fluids.

l. P.R. Berman and J. Ziegler, "Generalized Dressed Atom Approach to Atom - Strong Field Interactions -- Application to the Theory of Lasers and Bloch-Siegert Shifts", Physical Review A15, 2052-2052 (1977).

m. P.R. Berman, "Study of Collision Effects by Laser Spectroscopy, to appear as a chapter in Volume 13 of Advances in Atomic and Molecular Physics (Academic Press, New York).

n. J. Ziegler and P.R. Berman, "Theory of High Intensity Laser", to appear in Physical Review A.

o. C. Brechignac, R. Vetter and P.R. Berman, "Influence of Collisions on Saturated Absorption Profiles of the 557nm Line of Kr I", submitted to Journal of Physics B.